

# Dynamical spin response of electron doped Mott insulators on a triangular lattice

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The spin dynamics of electron doped Mott insulators on a triangular lattice is studied based on the  $t$ - $J$  model. It is found that the particularly universal behaviors of integrated dynamical spin structure factor seen in the doped Mott insulators on a square lattice, are absent in the doped Mott insulators on a triangular lattice, indicating the presence of the normal state gap. As a result, the spin-lattice relaxation rate  $1/T_1$  divided by  $T$  reduces with decreasing temperatures in the temperature region above  $0.2J \approx 50\text{K}$ , then follows a Curie-Weiss-like behavior at the temperature less than  $50\text{K}$ , in qualitative agreement with experimental observations.

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## I. INTRODUCTION

In the past few years, the discovery of superconductivity in layered cobalt oxides  $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ <sup>2</sup> has attracted considerable attention among the condensed matter physics society. Although the superconducting transition temperature ( $T_c \approx 4.5\text{K}$ )<sup>2</sup> is very low, the layered electronic structure and the dome-shaped behavior of  $T_c$  suggest this hydrated compound may be another doped Mott insulator<sup>2,3</sup>. In contrast to the cuprate superconductors with a square lattice of  $\text{CuO}_2$  planes, the parent compound  $\text{Na}_x\text{CoO}_2$  is composed of two-dimensional (2D)  $\text{CoO}_2$  layer with triangular geometry. This geometry frustration results in some novel electronic and magnetic phases, for instance, the Anderson's resonating valence state<sup>4</sup> and the strong topological frustration phases<sup>5,6</sup>. With the doping variation,  $\text{Na}_x\text{CoO}_2$  displays rich and complicated phase diagram<sup>7</sup>. In particular, superconductivity appears around  $x = 0.3$  when water molecules are intercalated between the  $\text{CoO}_2$  layers<sup>2,3</sup>.

Although extensive researches on this compound have been done in both experimental and theoretical sides, the doping dependent magnetic properties still remain unclear. The local spin density approach<sup>8</sup> predicted ferromagnetic coupling within 2D  $\text{CoO}_2$  layer for nearly all electron doping  $0.3 \leq x \leq 0.7$ . In fact, up to now, only at large doping neutron scattering studies and time-of-flight experiments on  $\text{Na}_{0.75}\text{CoO}_2$  and  $\text{Na}_{0.82}\text{CoO}_2$ <sup>9,10</sup> respectively, present low-energy fluctuations characteristic of A-type antiferromagnetism: antiferromagnetically coupled ferromagnetic layers. However, with the decreasing of Na content, a series of nuclear magnetic resonance (NMR)<sup>11</sup> and nuclear quadrupole resonance (NQR)<sup>12</sup> as well as Knight shift measurement<sup>13</sup> show the presence of strong 2D antiferromagnetic (AF) spin correlations and suggest the possibility of the spin-singlet superconductivity. That is to say the 2D AF spin correlations may be realized in the region of small  $x$  value for  $\text{Na}_x\text{CoO}_2$ . Starting from ab-initio band structure calculations, Korshunov and co-workers have estimated a certain critical concentration  $x_m$ , below which the magnetic susceptibility within the  $\text{CoO}_2$  plane shows a tendency towards

AF fluctuations<sup>14</sup>. The recent correlation effects on the doped  $\text{Na}_x\text{CoO}_2$  via a cellular cluster approach has reconfirmed and explained such in-plane magnetic transition from FM tendencies towards to AF<sup>15</sup>.

Recently, the surface-sensitive angle resolved photo emission spectroscopy (ARPES) experiments<sup>16–19</sup> observe a doping-dependent evolution of the fermi surface, which is found to be centered around the  $\Gamma$  point with a mostly  $a_{1g}$  character and no sign of six pockets resulting from the  $e'_g$  band predicted by local density approximation calculation<sup>8</sup> for a wide of Na concentrations. This indicates the importance of the electronic corrections in the doped  $\text{Na}_x\text{CoO}_2$ . Therefore, as discussed by many researchers<sup>5,6,20,21</sup>, an effective single band low-energy model suffices to capture the essential physics of electron-doped cobaltate  $\text{Na}_x\text{CoO}_2$ . With this consideration in mind, in this paper we also take a single band  $t$ - $J$  model as our starting point to discuss its unusual dynamical spin response. Since the AF fluctuations within the  $\text{CoO}_2$  planes may be realized in the low electron doping range, we are focusing on such region in our calculations below. We find that the particularly universal behaviors of integrated dynamical spin structure factor seen in the doped cuprates, are absent in the doped cobaltates, indicating the presence of the normal state gap. We also find that the spin-lattice relaxation rate  $1/T_1$  divided by  $T$  reduces with decreasing temperatures in the temperature region above  $0.2J \approx 50\text{K}$ , then follows a Curie-Weiss-like behavior at the temperature less than  $50\text{K}$  originating from the strong AF fluctuations, in qualitative agreement with experimental observations<sup>11,12</sup>. Our results also show that AF fluctuations within the  $\text{CoO}_2$  plane gradually weaken with increasing electron doping.

## II. THE $t$ - $J$ MODEL AND FERMION-SPIN THEORY

It has been argued that the essential physics of the doped  $\text{CoO}_2$  plane is contained in the  $t$ - $J$  model on a

triangular lattice<sup>5</sup>,

$$H = -t_e \sum_{i\hat{\eta}\sigma} PC_{i\sigma}^\dagger C_{i+\hat{\eta}\sigma} P^\dagger - \mu \sum_{i\sigma} PC_{i\sigma}^\dagger C_{i\sigma} P^\dagger + J \sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}}, \quad (1)$$

where the summation is over all sites  $i$ , and for each  $i$ , over its nearest-neighbor  $\hat{\eta}$ ,  $C_{i\sigma}^\dagger$  ( $C_{i\sigma}$ ) is the electron creation (annihilation) operator,  $\mathbf{S}_i = C_{i\sigma}^\dagger \boldsymbol{\sigma} C_{i\sigma} / 2$  is the spin operator with  $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$  as the Pauli matrices,  $\mu$  is the chemical potential, and the projection operator  $P$  removes zero occupancy, i.e.,  $\sum_\sigma C_{i\sigma}^\dagger C_{i\sigma} \geq 1$ . In the past fifteen years, some useful methods have been proposed to treat the no double occupancy local constraint in hole doped cuprates. In particular, a fermion-spin theory based on the partial charge-spin separation has been developed to study the physical properties of doped cuprates<sup>22,23</sup>, where the no double occupancy local constraint can be treated properly in analytical calculations. To apply this theory in the electron doped cobaltates, the  $t$ - $J$  model (1) can be rewritten in terms of a particle-hole transformation  $C_{i\sigma} \rightarrow f_{i-\sigma}^\dagger$  as,

$$H = -t \sum_{i\hat{\eta}\sigma} f_{i\sigma}^\dagger f_{i+\hat{\eta}\sigma} + \mu \sum_{i\sigma} f_{i\sigma}^\dagger f_{i\sigma} + J \sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}}, \quad (2)$$

supplemented by the local constraint  $\sum_\sigma f_{i\sigma}^\dagger f_{i\sigma} \leq 1$  to remove double occupancy, where for convenience, we have set  $t = -t_e < 0$ ,  $f_{i\sigma}^\dagger$  ( $f_{i\sigma}$ ) is the hole creation (annihilation) operator. Then the hole operators can be decoupled as,  $f_{i\uparrow} = a_{i\uparrow}^\dagger S_i^-$  and  $f_{i\downarrow} = a_{i\downarrow}^\dagger S_i^+$ , in the charge-spin separation fermion-spin theory<sup>22,23</sup>, where the spinful fermion operator  $a_{i\sigma} = e^{-i\Phi_{i\sigma}} a_i$  describes the charge degree of freedom together with some effects of the spin configuration rearrangements due to the presence of the doped electron itself (charge carrier), while the spin operator  $S_i$  describes the spin degree of freedom (spin), then the single occupancy local constraint,  $\sum_\sigma f_{i\sigma}^\dagger f_{i\sigma} = S_i^+ a_{i\uparrow}^\dagger a_{i\uparrow} S_i^- + S_i^- a_{i\downarrow}^\dagger a_{i\downarrow} S_i^+ = a_i a_i^\dagger (S_i^+ S_i^- + S_i^- S_i^+) = 1 - a_i^\dagger a_i \leq 1$ , is satisfied in analytical calculations. In this charge-spin separation fermion-spin representation, the low-energy behavior of the  $t$ - $J$  model (2) can be expressed as<sup>24</sup>,

$$H = -t \sum_{i\hat{\eta}} (a_{i\uparrow} S_i^+ a_{i+\hat{\eta}\uparrow}^\dagger S_{i+\hat{\eta}}^- + a_{i\downarrow} S_i^- a_{i+\hat{\eta}\downarrow}^\dagger S_{i+\hat{\eta}}^+) - \mu \sum_{i\sigma} a_{i\sigma}^\dagger a_{i\sigma} + J_{\text{eff}} \sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}}, \quad (3)$$

with  $J_{\text{eff}} = (1-x)^2 J$ , and  $x = \langle a_{i\sigma}^\dagger a_{i\sigma} \rangle = \langle a_i^\dagger a_i \rangle$  is the electron doping concentration. In this case, the magnetic energy ( $J$ ) term in the  $t$ - $J$  model is only to form an adequate spin configuration, while the kinetic energy ( $t$ ) term has been transferred to the charge carrier-spin interaction, which dominates the essential physics.

In the framework of the charge-spin separation, the basic low-energy excitations are charge carriers and spins. It has been shown that the charge dynamics is mainly governed by the scattering from the charge carriers due to the spin fluctuation<sup>24</sup>, while the spin fluctuations couple only to the spin and therefore no composition law is required in discussing the spin dynamics<sup>25</sup>, but the effect of charge carriers is still considered through the charge carrier's order parameter  $\phi$  entering the spin propagator. In this case, the spin dynamics of the doped square antiferromagnet has been discussed<sup>25</sup> by considering the spin fluctuation around the mean-field solution, where the spin part is treated by the loop expansion to the second order. Following their discussions, we can obtain the dynamical spin structure for the electron doped cobaltates as

$$S(k, \omega) = \text{Re} \int_0^\infty dt e^{i\omega(t-t')} D(k, t-t') = -2[1 + n_B(\omega)] \text{Im} D(k, \omega), \quad (4)$$

respectively, where the full spin Green's function,  $D^{-1}(k, \omega) = D^{(0)-1}(k, \omega) - \Sigma_s(k, \omega)$ , with the mean-field (MF) spin Green's functions  $D^{(0)-1}(k, \omega) = (\omega^2 - \omega_k^2)/B_k$ , and the second-order spin self-energies from the charge carrier pair bubble,

$$\Sigma_s(k, \omega) = \left(\frac{Zt}{N}\right)^2 \sum_{pp'} (\gamma_{p'+p+k}^2 + \gamma_{p'-k}^2) \frac{B_{k+p}}{2\omega_{k+p}} \times \left( \frac{F_1^{(s)}(k, p, p')}{\omega + \xi_{p+p'} - \xi_{p'} - \omega_{k+p}} - \frac{F_2^{(s)}(k, p, p')}{\omega + \xi_{p+p'} - \xi_{p'} + \omega_{k+p}} \right) \quad (5)$$

where  $Z$  is the number of the nearest neighbor sites,  $B_k = \lambda[2\chi^z(\epsilon\gamma_k - 1) + \chi(\gamma_k - \epsilon)]$ ,  $\lambda = 2ZJ_{\text{eff}}$ ,  $\epsilon = 1 + 2t\phi/J_{\text{eff}}$ ,  $\gamma_k = [\cos k_x + 2\cos(k_x/2)\cos(\sqrt{3}k_y/2)]/3$ , the spin correlation function  $\chi^z = \langle S_i^z S_{i+\hat{\eta}}^z \rangle$ ,  $F_1^{(s)}(k, p, p') = n_F(\xi_{p+p'})[1 - n_F(\xi_{p'})] - n_B(\omega_{k+p})[n_F(\xi_{p'}) - n_F(\xi_{p+p'})]$ ,  $F_2^{(s)}(k, p, p') = n_F(\xi_{p+p'})[1 - n_F(\xi_{p'})] + [1 + n_B(\omega_{k+p})][n_F(\xi_{p'}) - n_F(\xi_{p+p'})]$ ,  $n_B(\omega_p)$  and  $n_F(\xi_p)$  are the boson and fermion distribution functions, respectively. The MF charge carrier and spin spectra are given by  $\xi_k = Zt\chi\gamma_k - \mu$ , and  $\omega_k^2 = A_1(\gamma_k)^2 + A_2\gamma_k + A_3$ , respectively, with  $A_1 = \alpha\epsilon\lambda^2(\epsilon\chi^z + \chi/2)$ ,  $A_2 = -\epsilon\lambda^2[\alpha(\chi^z + \epsilon\chi/2) + (\alpha C^z + (1-\alpha)/(4Z) - \alpha\epsilon\chi/(2Z)) + (\alpha C + (1-\alpha)/(2Z) - \alpha\chi^z/2)/2]$ ,  $A_3 = \lambda^2[\alpha C^z + (1-\alpha)/(4Z) - \alpha\epsilon\chi/(2Z) + \epsilon^2(\alpha C + (1-\alpha)/(2Z) - \alpha\chi^z/2)/2]$ , and the spin correlation function  $\chi = \langle S_i^+ S_{i+\hat{\eta}}^- \rangle$ ,  $C = (1/Z^2) \sum_{\hat{\eta}\hat{\eta}'} \langle S_{i+\hat{\eta}}^+ S_{i+\hat{\eta}'}^- \rangle$ , and  $C^z = (1/Z^2) \sum_{\hat{\eta}, \hat{\eta}'} \langle S_{i+\hat{\eta}}^z S_{i+\hat{\eta}'}^z \rangle$ . In order not to violate the sum rule of the correlation function  $\langle S_i^+ S_i^- \rangle = 1/2$  in the case without AF long-range order, the important decoupling parameter  $\alpha$  has been introduced in the MF calculation<sup>26,27</sup>, which can be regarded as a vertex correction. All the above MF order parameters, decoupling parameter  $\alpha$ , and chemical potential  $\mu$  are determined by the self-consistent calculation<sup>27</sup>.

### III. NUMERICAL RESULTS AND DISCUSSIONS

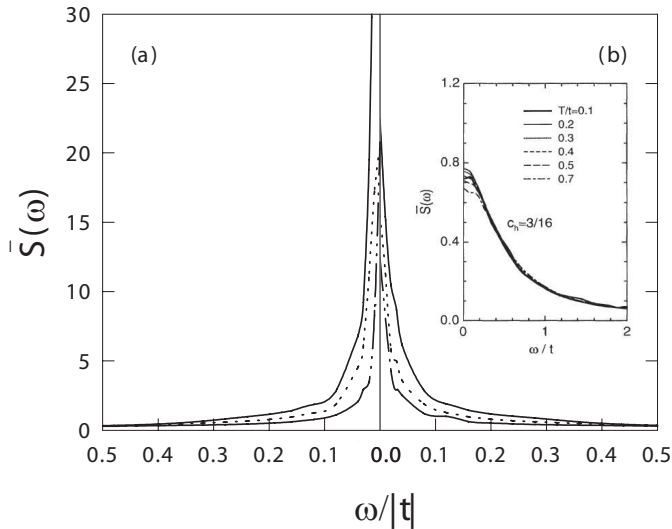


FIG. 1: The integrated dynamical spin structure factor at the doping (a)  $x = 0.33$  and (b)  $x = 0.30$  with the temperature  $T = 0.3J$  (dashed-dotted line),  $T = 0.4J$  (dotted line), and  $T = 0.6J$  (solid line) for the parameter  $t/J = -2.5$ . Inset: the numerical simulation taken from Ref. [27].

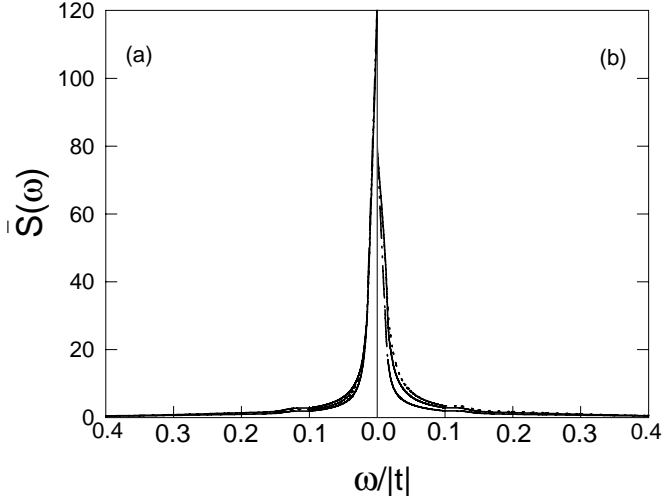


FIG. 2: The integrated dynamical spin structure factor at the doping (a)  $x = 0.33$  and (b)  $x = 0.30$  with the temperature  $T = 0.7J$  (dashed-dotted line),  $T = 0.8J$  (dotted line), and  $T = 0.9J$  (solid line) for the parameter  $t/J = -2.5$ .

For understanding the dynamic spin response of the electron doped cobaltates, we have calculated the integrated dynamical spin structure factor, which can be expressed as

$$\bar{S}(\omega, T) = S_L(\omega) + S_L(-\omega) = (1 + e^{-\beta\omega})S_L(\omega)$$

$$= (1 + e^{-\beta\omega}) \frac{1}{N} \sum_k S(k, \omega). \quad (6)$$

The numerical results of the integrated dynamical spin structure factor at doping (a)  $x = 0.33$  and (b)  $x = 0.30$  with the temperature  $T = 0.3J$  (dashed-dotted line),  $T = 0.4J$  (dotted line), and  $T = 0.6J$  (solid line) for the parameter  $t/J = -2.5$  are plotted in Fig. 1. It is shown that the integrated dynamical spin structure factor decreases with increasing energies for  $\omega < 0.3t$ , and almost constant for  $\omega \geq 0.3t$ . which is similar to the case of doped cuprates<sup>25,28</sup> and reflects the common features of the doped Mott insulators. However, the temperature dependence of the integrated dynamical spin structure factor, which is essentially temperature independent at high energy in a wide temperature regime, presents a strong deviation at the low-energy range, so that the shape of the integrated dynamical spin structure factor does not follow a universal behavior, in contrast to the case of the doped cuprates (seen the insert in Fig. 1b)<sup>28</sup>. It has been discussed that the universal behavior of the integrated dynamical spin structure factor in the doped cuprates is due to the absence of the normal state gap in the electron density of states<sup>25</sup>. However, in the doped cobaltates, the normal state gap opens<sup>11,29,30</sup>, which is mainly induced by the frustrated spin. Since in the charge-spin separation framework the spin dynamics is dominated by the scattering of spins, which are strongly renormalized because of the strong interactions with fluctuations of surrounding charge carrier excitations. The frustrated spin moves in the background of the charge carriers, and the cloud of distorted charge carriers background is to follow the frustrated spins, which leads to the anomalous spin dynamics in the doped cobaltates. In other words, the origin of the absence of the particularly universal behavior of the integrated dynamical spin structure factor in the doped cobaltates at low energy originates from the normal-state gap due to the magnetic frustration. In order to show this normal state gap clearly, we calculate the  $\bar{S}(\omega, T)$  at higher temperature as shown in Fig. 2, where the parameters are the same with the Fig. 1 except for the temperature  $T = 0.7J$  (dashed-dotted line),  $T = 0.8J$  (dotted line), and  $T = 0.9J$  (solid line). In Fig. 2, the temperature dependent integrated dynamical spin structure factor nearly degenerates and doesn't show a deviation at low energy seen in Fig. 1. This universal behavior of the integrated dynamical spin structure factor exists for the whole temperature range is similar to the case of doped cuprates (seen the insert in Fig. 1b)<sup>25,28</sup>. From Fig. 1 and Fig. 2, we estimate the normal state gap opens at temperature  $T_{gap} \simeq 0.7J$ . According to our estimation  $J \simeq 250K$ <sup>6</sup>, we get the  $T_{gap} \simeq 160K$ , very close to the experiments<sup>11</sup>.

Now we turn our attention to discuss the spin-lattice relaxation rate  $1/T_1$ . Theoretically, the spin contributions to the spin-lattice relaxation rate  $1/T_1$  divided by  $T$  may be written using the imaginary part of the Co

dynamical electron spin-susceptibility  $\chi''(\mathbf{k}, \omega)$  as

$$\frac{1}{T_1 T} = \frac{2\gamma_i^2 k_B}{g^2 \mu_B^2} \lim_{\omega \rightarrow 0} \sum_k |A(k)|^2 \frac{\chi''(\mathbf{k}, \omega)}{\omega}, \quad (7)$$

where  $\chi''(k, \omega) = [1 + n_B(\omega)]^{-1} S(k, \omega)$ ,  $g$  is the  $g$  factor,  $\mu_B$  is the Bohr magneton, and  $A(k) = \sum_i A_i e^{ikr_i}$ , where  $A_i$  is the hyperfine coupling between the electron spin and the nuclear spin having gyromagnetic ratio  $\gamma_i$ <sup>31</sup> and is to be approximated to a constant in our calculation. The

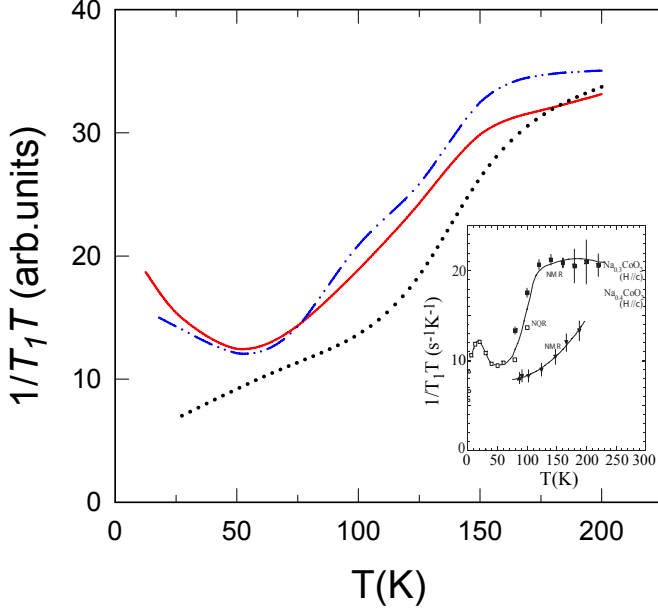


FIG. 3: (color online) The spin-lattice relaxation rate  $1/T_1$  divided by  $T$  of the electron doped cobaltates as a function of temperature at doping  $x = 0.30$  (solid line),  $x = 0.33$  (dashed-dotted line), and  $x = 0.40$  (dotted line) for parameter  $t/J = -2.5$ , where we choose  $2\gamma_i^2 k_B A_i / g^2 \mu_B^2 = \text{constant}$ . Inset: the experimental result taken from Ref. [10].

spin-lattice relaxation rate  $1/T_1$  divided by  $T$  in Eq.(7) has been evaluated numerically and the results for  $t/J = -2.5$  at the doping  $x=0.30$  (solid line),  $x=0.33$  (dashed-dotted line), and  $x=0.40$  (dotted line) in comparison with the corresponding experiments (inset) are plotted in Fig. 3. It is shown that at low doping  $x=0.30$  (solid line) and  $x=0.33$  (dashed-dotted line), the spin-lattice relaxation rate  $1/T_1 T$  reduces with decreasing temperature in the temperature region  $0.2J \approx 50K < T < 0.7J \approx 160K$ , which is self-consistent with the integrated dynamical spin susceptibility shown in Fig. 1 and Fig. 2, and is the result of the opening of normal state gap. However, in the low temperature region  $T < 0.2J$ , the spin-lattice

relaxation rate  $1/T_1 T$  follows a Curie-Weiss-like behavior due to the existence of strong AF fluctuations. However, at the doping  $x=0.40$  (dotted line), although the normal state gap still exists at high temperature, Curie-Weiss-like behavior at low temperature disappears, indicating that AF-fluctuations have become weak with the increasing doping. These results are in good agreement with the recent experiments<sup>11,12</sup>. At the high doping  $x > 0.4$ , the neutron scattering studies and time-of-flight experiments on  $\text{Na}_{0.75}\text{CoO}_2$  and  $\text{Na}_{0.82}\text{CoO}_2$ <sup>9,10</sup>, respectively, have observed the ferromagnetic correlation in the 2D  $\text{CoO}_2$  plane. Therefore, the magnetic fluctuation in the 2D  $\text{CoO}_2$  plane has been affected by the electron doping, *i.e.*, the magnetic fluctuation is doping dependent. The later ab-initio band structure calculations for  $\text{Na}_x\text{CoO}_2$  have given a certain critical value  $x_m = 0.56 \sim 0.68$ . The magnetic susceptibility in the  $\text{CoO}_2$  plane shows a tendency towards the AF state below  $x_m$  and ferromagnetic state above  $x_m$ <sup>14</sup>.

#### IV. SUMMARY

In summary, we have studied the dynamical spin response in the electron doped cobaltates within the  $t$ - $J$  model. We have shown that due to the presence of normal state gap, the particularly universal behaviors of integrated dynamical spin structure factor seen in the doped cuprates at the low energy, are absent in the doped Mott insulators on a triangular lattice. We have also shown that the spin-lattice relaxation rate  $1/T_1$  divided by  $T$  reduces with decreasing temperature in the temperature region above  $0.2J \approx 50K$ , then follows a Curie-Weiss-like behavior at the temperature less than  $50K$  originating from the strong antiferromagnetic fluctuations, in qualitative agreement with experiments. Our results also indicate that the electron doping plays a important role in altering the magnetic properties of the  $\text{CoO}_2$  plane.

#### V. ACKNOWLEDGMENTS

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